INL/EXT-13-29948

PINS Variance Study

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ABSTRACT

The INL's Portable Isotopic Neutron Spectroscopy System (PINS)[1] non-intrusively identifies the chemical fill of munitions and sealed containers. PINS is used routinely by the U.S. Army, the Defense Threat Reduction Agency, and foreign military units to determine the contents of munitions and other containers suspected to contain explosives, smoke-generating chemicals, and chemical warfare agents such as mustard and nerve gas. The objects assayed with PINS range from softball-sized M139 chemical bomblets to 200 gallon DOT 500X ton containers.

INL had previously examined[2] the feasibility of using a similar system for the identification of explosives, and based on this proof-of-principle test, the development of a dedicated system for the identification of explosives in the presence of special nuclear material for emergency responders was started in 2011. INL tested prototype PINS on explosives and mock explosives with kilogram quantities of highly-enriched uranium and weapons-grade plutonium in 2011. In 2012 INL concentrated on making the system more sensitive to identifying explosives, particularly in the detection of the nitrogen content of explosives. This past year INL has been concentrating its efforts in studying the variance in the spectra, in particular the variance in the elemental ratios that are used to identify individual explosives.

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ACRONYMS

DT	Deuterium tritium
HEU	Highly enriched uranium
HPGe	High purity germanium
INL	Idaho National Laboratory
MCA	Multi channel analyzer
MCNP	Monte Carlo n-particle
NDSTC	Nuclear Detection and Sensor Testing Center
NNSA	National Nuclear Security Administration
PINS	Portable Isotopic Neutron Spectroscopy
SNM	Special nuclear material
ZPPR	Zero Power Physics Reactor

PINS Explosives Identification

1. INTRODUCTION

PINS employs neutron radiation to probe the chemical elements within a container without the need to open or even touch the container. A radioisotopic neutron source or an electronic neutron generator shines neutrons on the item under test. The neutrons, in turn, penetrate the container or munition where they interact with the atomic nuclei of the filler material, producing gamma rays characteristic of the chemical elements inside the item. These energetic gamma rays can penetrate even the thick steel wall of an artillery projectile to escape and trigger a gamma-ray spectrometer.

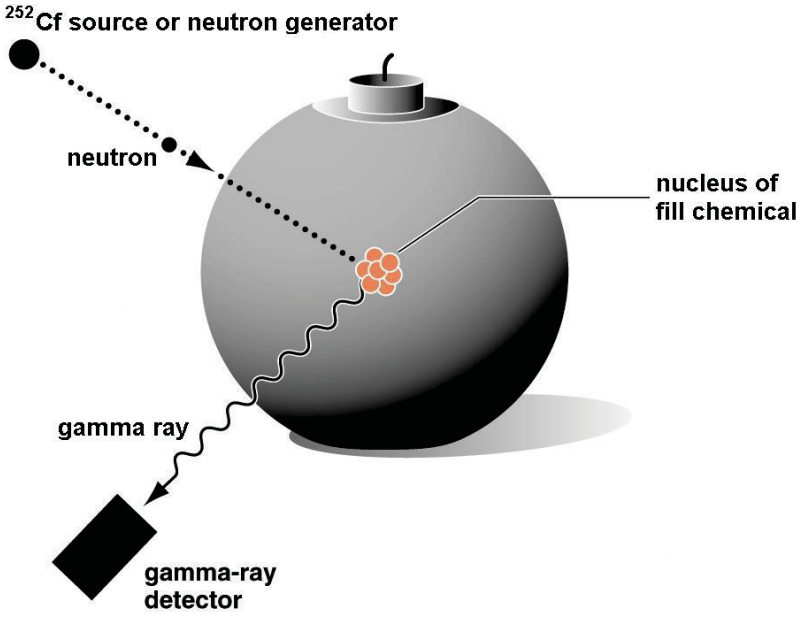

Figure 1: Schematic neutron reaction

This technique is called Prompt Gamma-ray Neutron Activation Analysis (PGNAA). A schematic neutron reaction event is shown above in Figure 1. The energies and intensities of these gamma rays, as measured by the spectrometer, identify the elemental composition of the fill material.

Table 1 Elemental composition[3] of explosives in weight percent

	HMX	LX-17	TNT	COMPB	TATB	PBX-9501	PBX-9502	PETN	LX-10-0	PBX-9404	TATP
H	2.7	2.2	2.2	2.7	2.3	2.9	2.2	2.55	2.65	2.8	8.2
C	16.2	27.5	37.0	24.3	27.9	17.7	27.6	19.0	17.4	16.8	48.6
O	43.2	34.4	42.3	42.6	37.2	43.0	35.3	60.7	41.3	36.0	43.2
N	37.8	30.1	18.5	30.4	32.6	36.4	31.0	17.7	36.0	43.0	
Cl		1.9					1.3			1.1	
P										0.3	
F		2.2					1.4		3.2		

Chemical Warfare Materiel (CWM), e.g. nerve agent GB, is relatively simple to identify based on its gamma-ray signature, due to the variety of chemical elements in the different agents. Explosives, however, are somewhat more difficult to distinguish from one another as they are largely comprised of the same four elements: carbon, hydrogen, nitrogen, and oxygen. Table 1 shows the elemental composition[3] of some explosives of interest. As can be seen in the table, some plastic-bonded explosives contain the elements chlorine , fluorine, or phosphorous, but the bulk of the material is still carbon, oxygen, hydrogen, and nitrogen.

Our previous work comprised studying the response of a gamma spectrometer to neutron-induced gamma rays from kilogram or larger masses of the explosives listed in Table 1 both in Monte Carlo simulations as well as a limited set of experiments. The conclusion from these studies was that using a deuterium-tritium (DT) neutron generator provided the best excitation of carbon and oxygen in the explosives and that the calculation of elemental ratios provided some discrimination of different explosive types.

Figure 2 below shows a plot of carbon-to-nitrogen and carbon-to-oxygen elemental ratios from MCNP calculations of the response of the HPGe detector in our system to various explosives. The gamma rays used in calculating these ratios are all due to neutron inelastic scattering of neutrons with energies greater than 4 MeV. Using gamma rays from neutron interactions based on similar neutron energies removes most of the effects from the shape of the explosives. As can be seen in the Figure, use of these elemental ratios provides a good degree of separation of the different explosive types. Areas where explosives overlap are largely due to explosives being based on the same type, e.g. PBX-9404, PBX-9501, and LX-10-0 are all HMX or RDX-based plastic-bonded explosives.

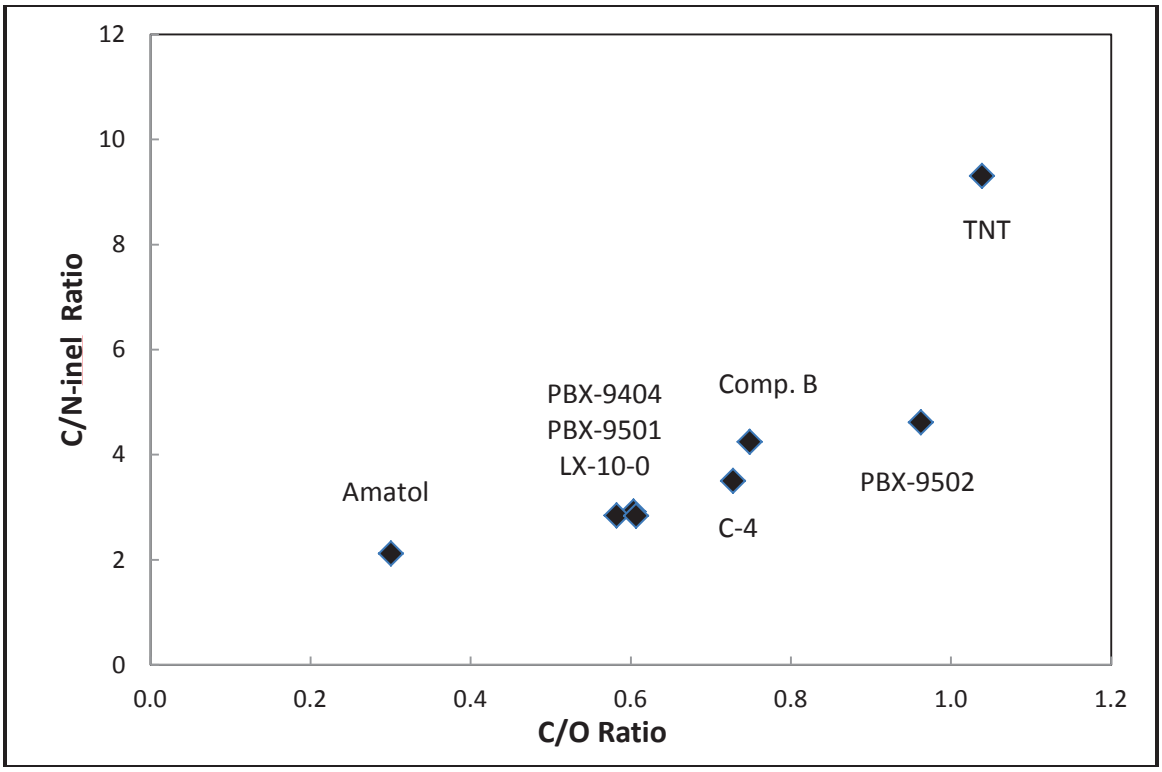

Figure 2: MCNP calculation of the peak area ratios for various explosives.

2. VARIANCE AS IT AFFECTS IDENTIFICATION

The use of elemental ratios to identify explosives requires that these ratios remain relatively invariant for a large range of geometries and masses of the explosives. In order to find the minimum variance that could be achieved a series of replicate measurements of both simulated explosives (simulants) and active background measurements were performed.

2.1 Background variance

The first measurement that is performed during a PINS assessment is an active background measurement. This measurement consists of running the system in the same configuration and location as that of the assessment of a test object, but with no test object present. This measurement allows any contribution to gamma-ray peaks of interest due to background materials such as the floor, shielding, or the detector itself to be determined. These contributions are then subtracted from the assessment spectra that are measured later. The experimental arrangement for a typical background measurement can be seen in Figure 3 below.

Figure 3. Typical background measurement.

As can be seen in the figure, one could reasonably expect to see contributions in the background gamma-ray spectrum from the aluminum frame, polyethylene moderator, neutron generator, and shielding

materials. If the peak areas from elements of interest such as carbon, hydrogen, oxygen, and nitrogen varied significantly in these background measurements, then the corresponding elemental ratios from assessments would also show variance after subtracting background even if the assessment peak areas did not have significant variance.

2.2 Elemental ratio variance

Replicate measurements of simulated explosives were measured for 2000 live seconds after each 1000 live second active background measurement. Six kilograms of simulated explosives in three two-liter Nalgene bottles were placed in the same position in front of the system for each assessment. An example of the experimental arrangement can be seen in Figure 4 below.

Figure 4. Typical simulant measurement.

Variance in the replicate measurements shows the limits to how well an individual explosive may be characterized. For example if the Comp. B C/O ratio typically varied by as much as 50% while also having a C/N ratio that typically varied by 50%, then Comp. B would frequently be mistakenly identified as PBX-9501 or PBX-9502 as one can see by examining Figure 2. If the variances in the Comp. B ratios are much smaller, then Comp. B can be reliably distinguished from other explosives.

3. REPLICATE MEASUREMENT RESULTS

The results of both the background and simulant measurements will be described in the sections below.

3.1 Replicate background measurements

Background measurements were made on multiple days with multiple detectors. These measurements were made for 1000 live seconds each. The peaks of interest were fit using the PINS+ data acquisition and analysis package in order to determine peak areas. An example of such a peak area can be seen in Figure 4 below where the carbon peak area for a set of replicate background measurements is plotted. These background measurements were measured on multiple days using two different detectors. The neutron generator was operated at the same nominal output for each of the measurements.

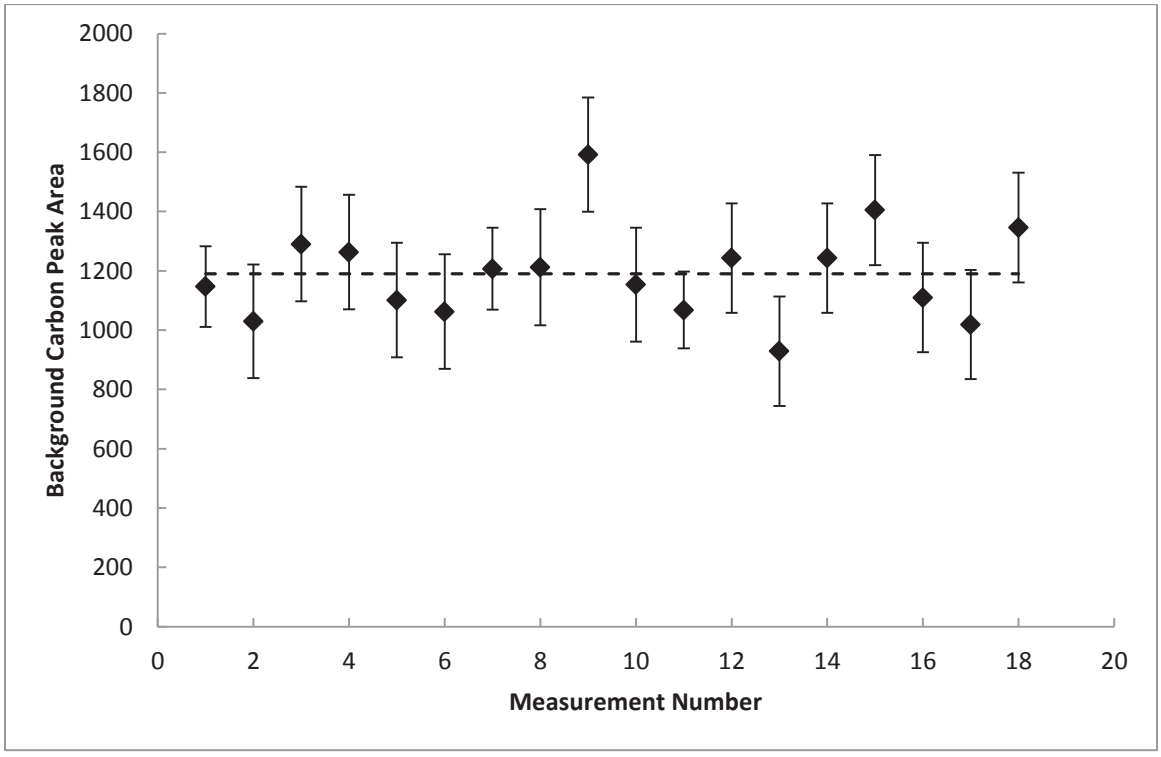

Figure 5. Carbon peak area for replicate background measurements

The figure shows that the individual carbon peak areas are in general within one standard deviation of the mean carbon peak area. The standard deviation of the mean for this data set is approximately 13%, which is significantly higher than one would expect from counting statistics. This is due to the method by which the carbon peak area is calculated.

The carbon peak, unlike the other peaks used in identification of explosives, is highly Doppler broadened as can be seen in Figure 6 below. Because of this broadening, it is difficult to fit using the same algorithms as are used for the other peaks which fit a gaussian to the peak location. Instead, two small regions, one lower in energy than the carbon peak and one higher are used to determine the level of the Compton continuum underneath the carbon peak. This continuum region is below the red line in Figure 6. Counts above this line are then assumed to be due to neutron inelastic scattering on carbon.

Part of the variance in the carbon peak area is likely due to variance in setting this Compton continuum level.

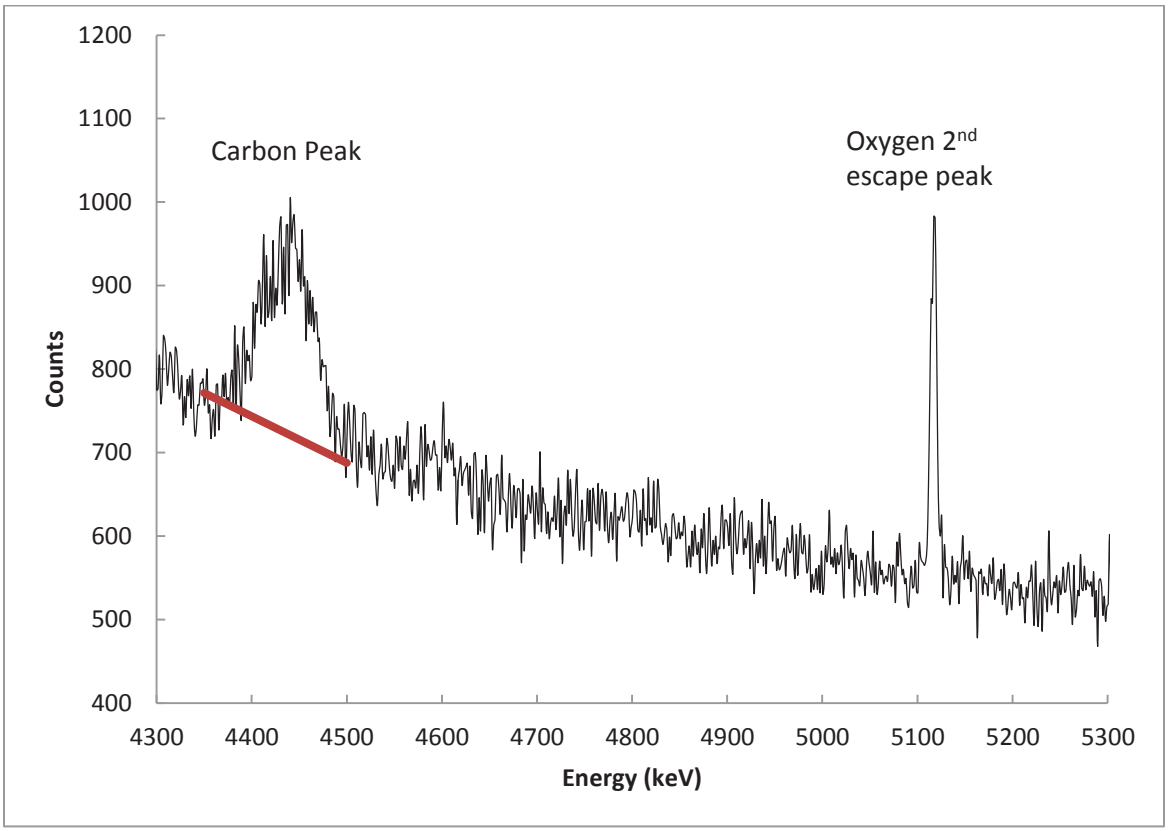

Figure 6. Carbon peak area for replicate background measurements

We see even greater variance in the oxygen, nitrogen, and hydrogen peak background peak areas as can be seen in Figure 7, Figure 8, and Figure 9 below. Some of this increased variance is expected due to the lower count rates in these three peaks, especially in the case of nitrogen. The nitrogen peak is also among these peaks in that its area is determined not just by fitting the region at 5106 keV where the nitrogen inelastic peak is located, but also by calculating the contribution to this area from the second escape peak of the oxygen full-energy peak at 6129 keV. Variance in the nitrogen peak area therefore includes contributions from the variance in the oxygen full-energy peak area.

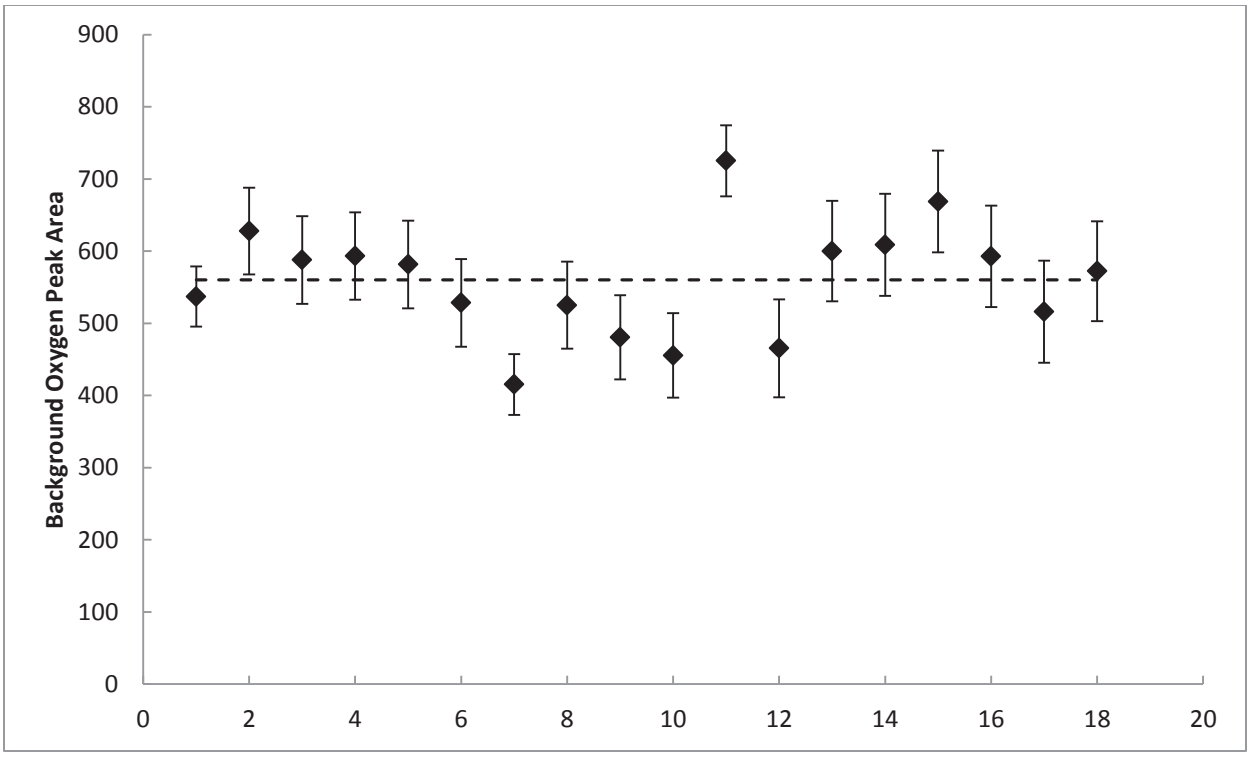

Figure 7. Oxygen peak area for replicate background measurements

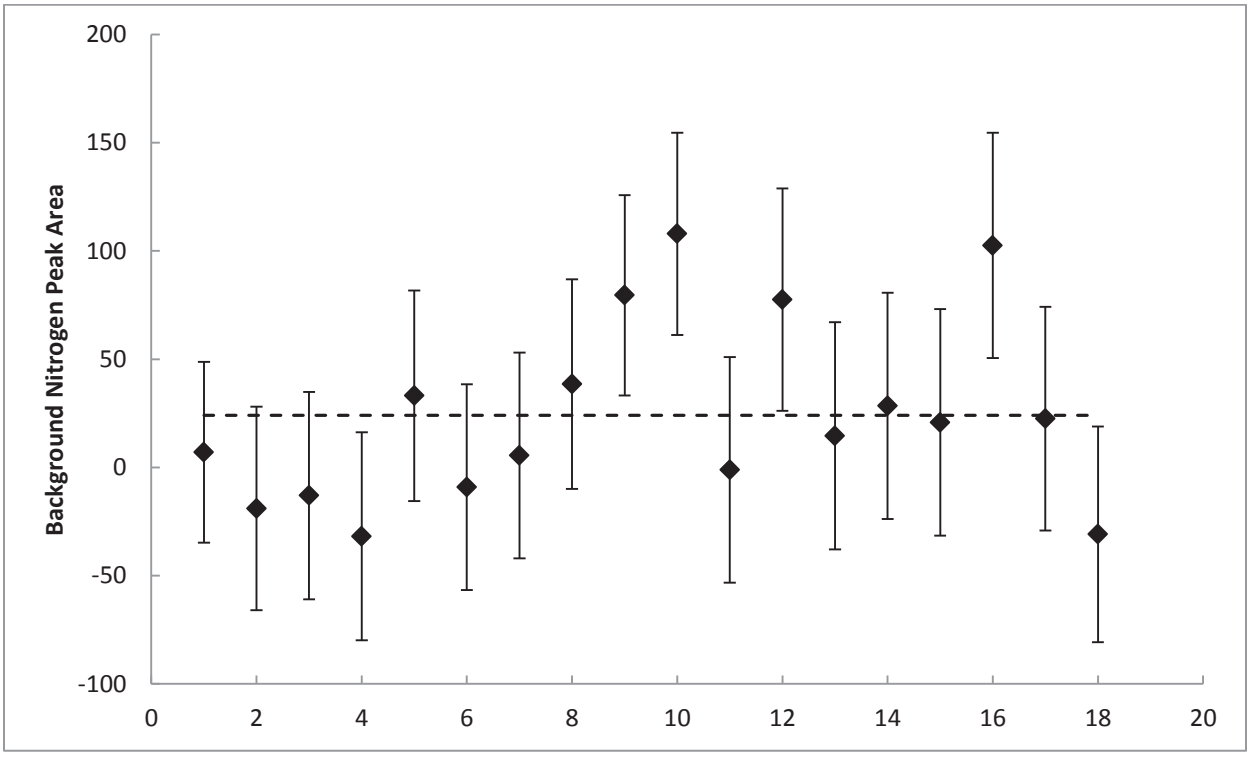

Figure 8. Nitrogen peak area for replicate background measurements

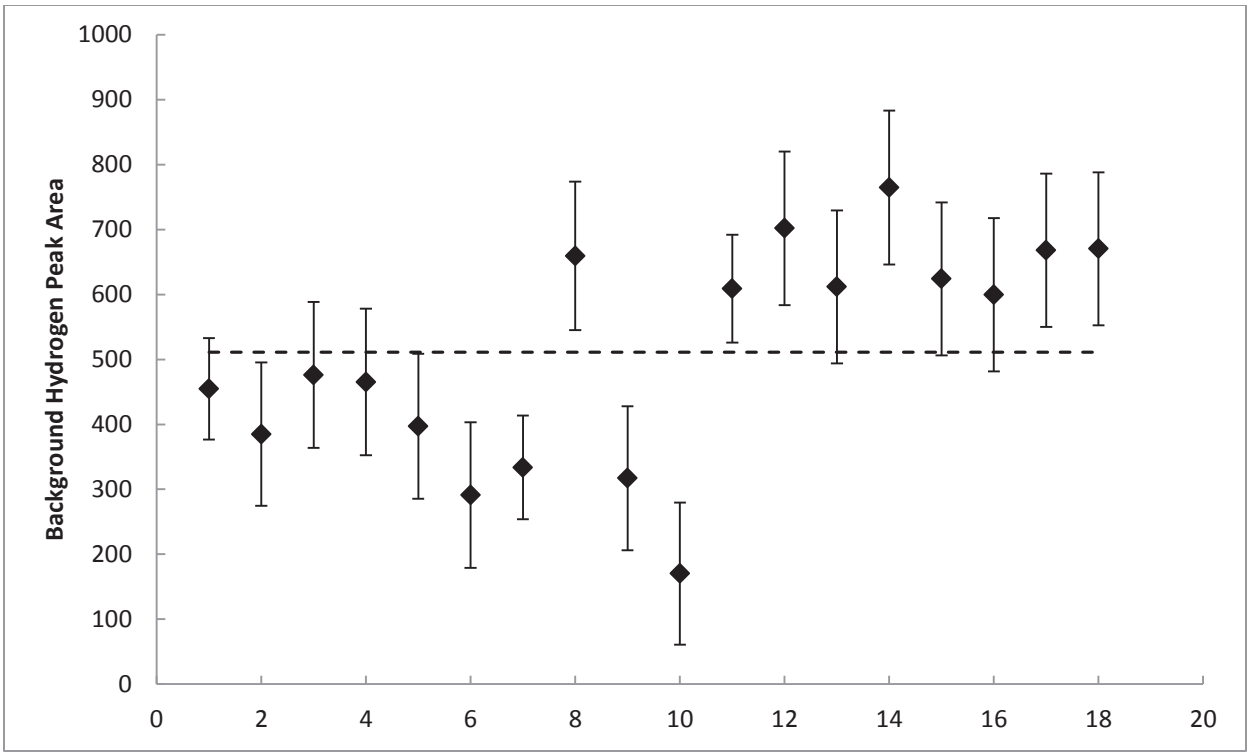

Figure 9. Hydrogen peak area for replicate background measurements.

3.2 Replicate Simulant Measurements

Three simulated explosives were used in these replicate measurements. Simulants for HMX, TNT, and TATB were used. These three simulants provide a range of elemental ratios large enough to show what impact significant variance would have on the identification of the explosives. Elemental ratios were measured for each of the three simulants and their variance was calculated. Elemental ratios for the HMX simulant can be seen in Figure 10, Figure 11, and Figure 12 below. In each of the plots, the standard deviation about the mean of the average elemental ratio is shown as two dashed lines.

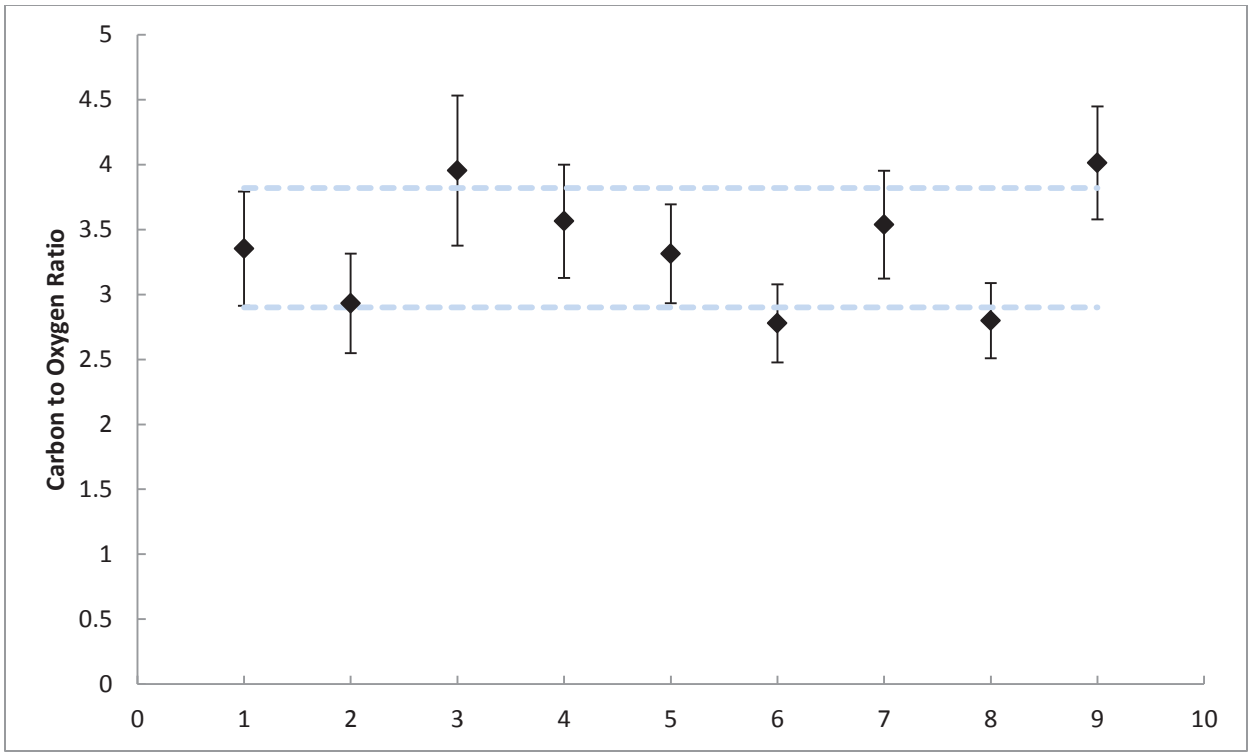

Figure 10. Carbon to oxygen ratio for simulated HMX.

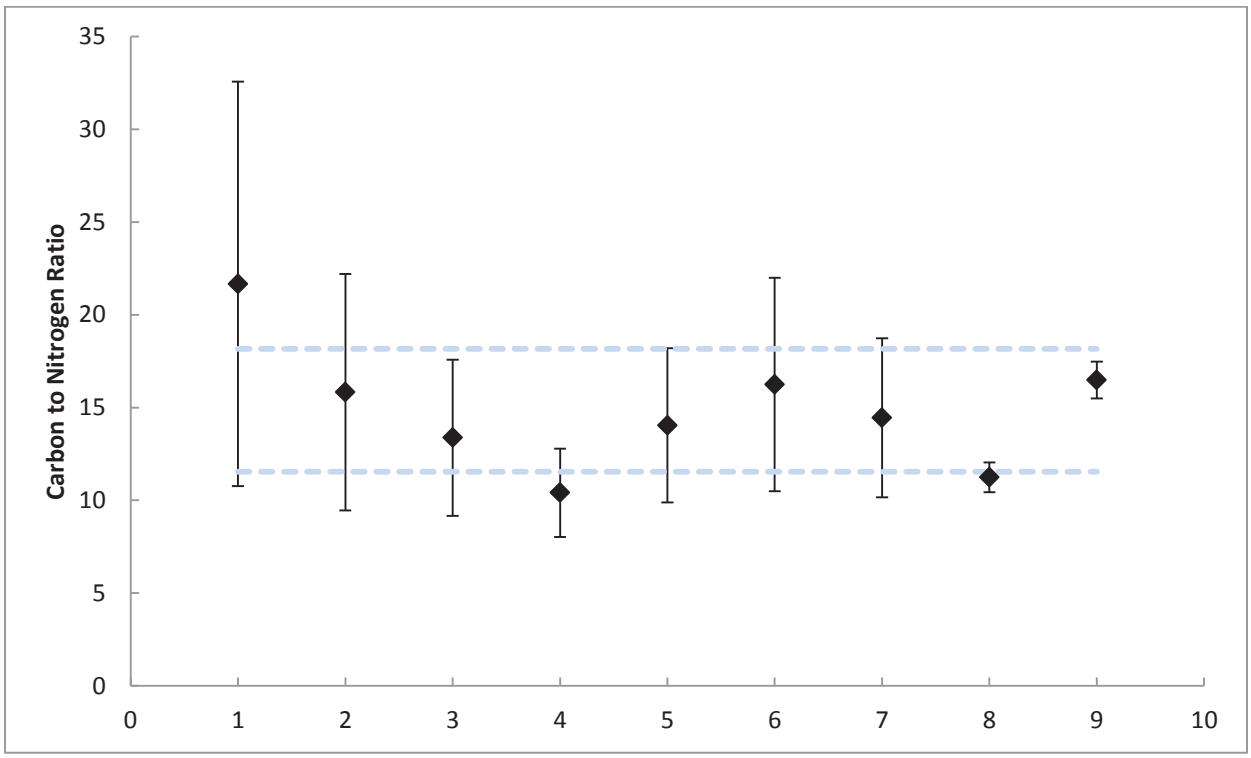

Figure 11. Carbon to nitrogen ratio for simulated HMX.

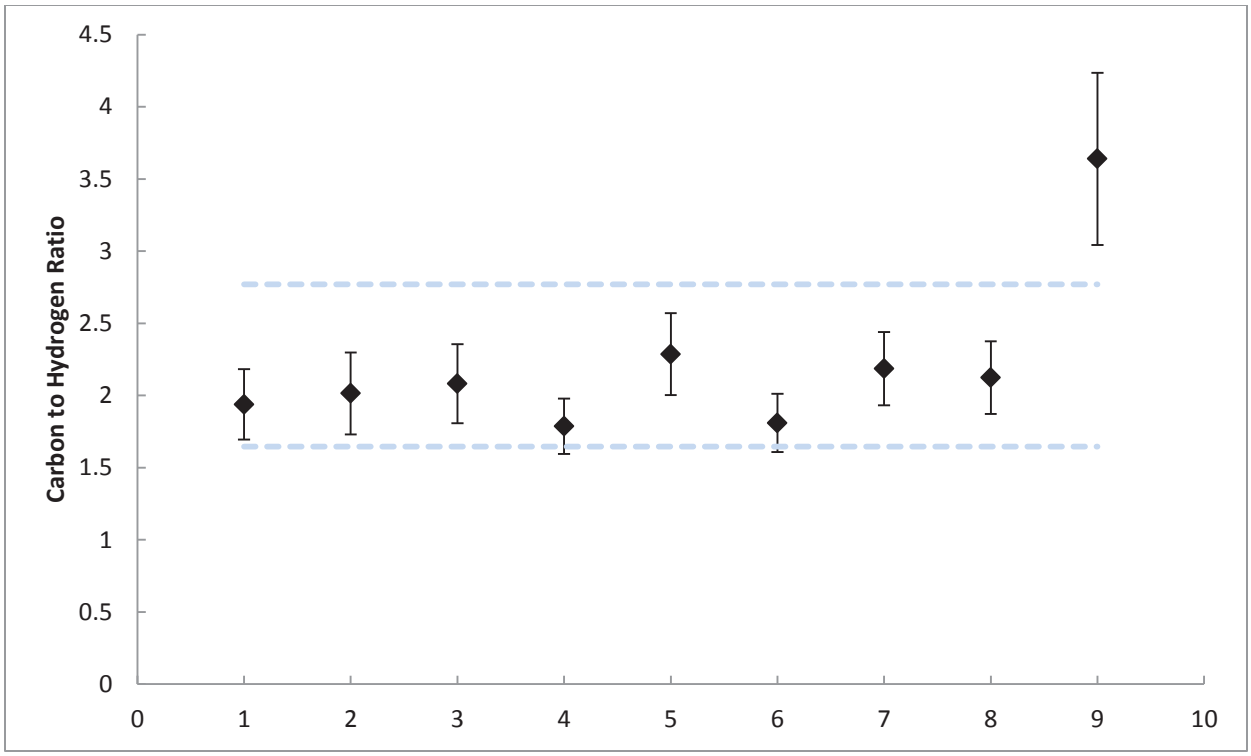

Figure 12. Carbon to hydrogen ratio for simulated HMX.

The same elemental ratios are shown below for the simulated TATB and TNT are shown in Figures 13-18 below.

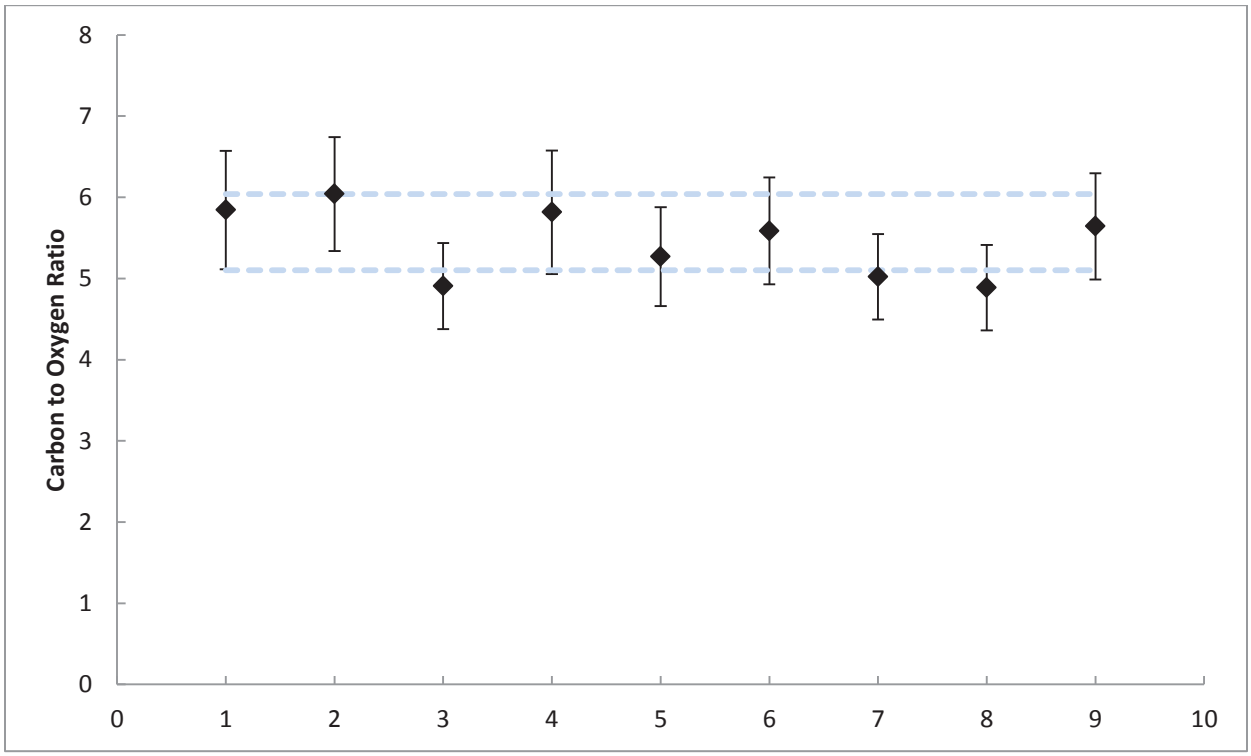

Figure 13. Carbon to oxygen ratio for simulated TATB.

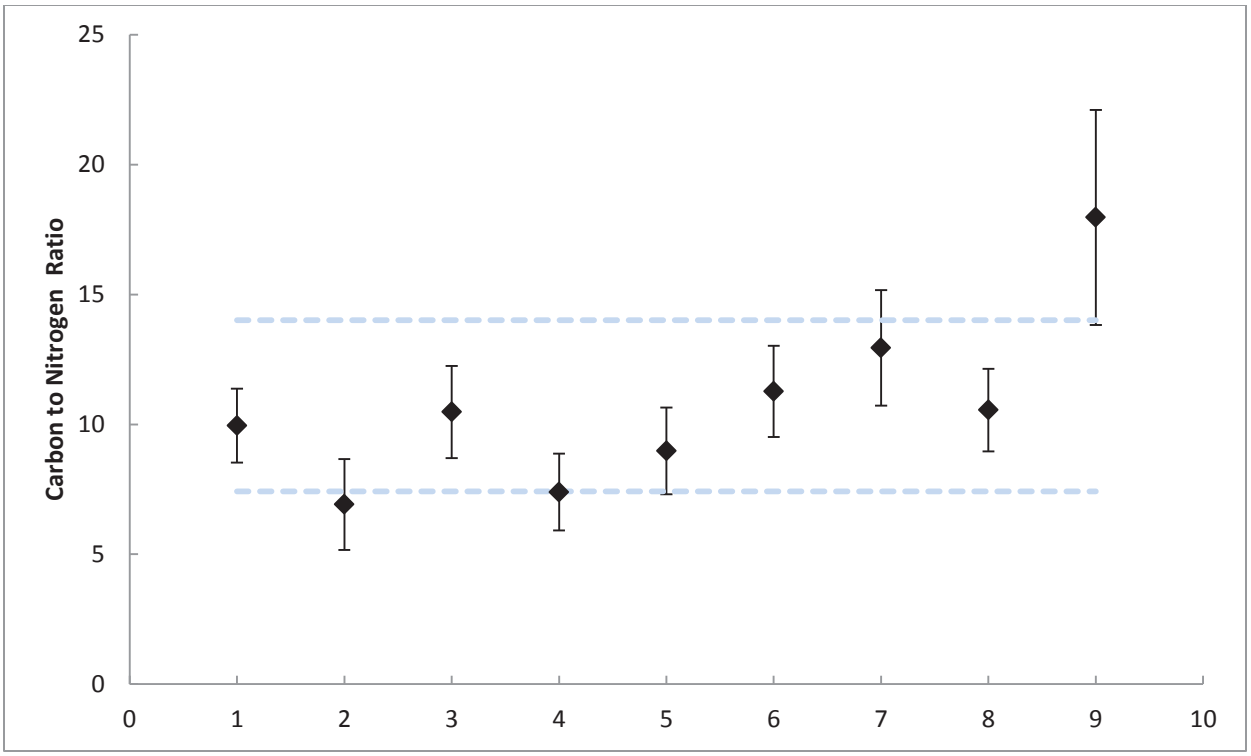

Figure 14. Carbon to nitrogen ratio for simulated TATB.

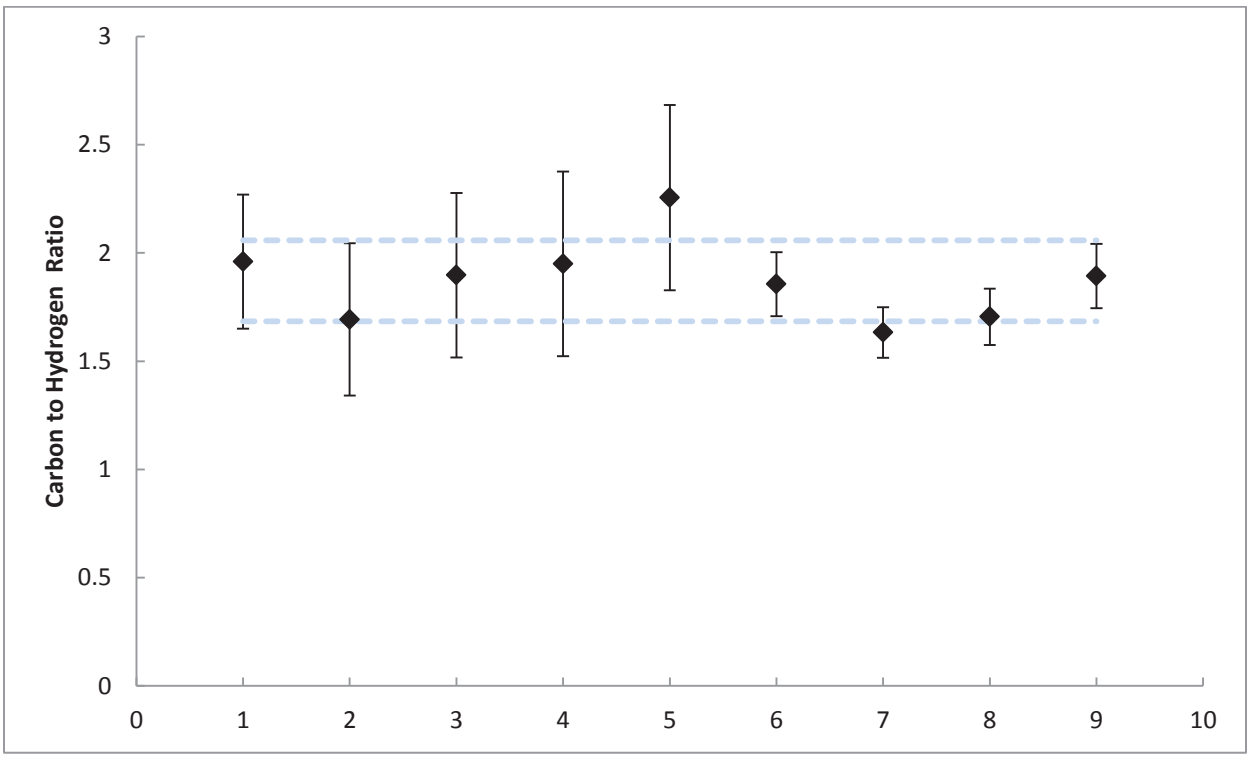

Figure 15. Carbon to hydrogen ratio for simulated TATB.

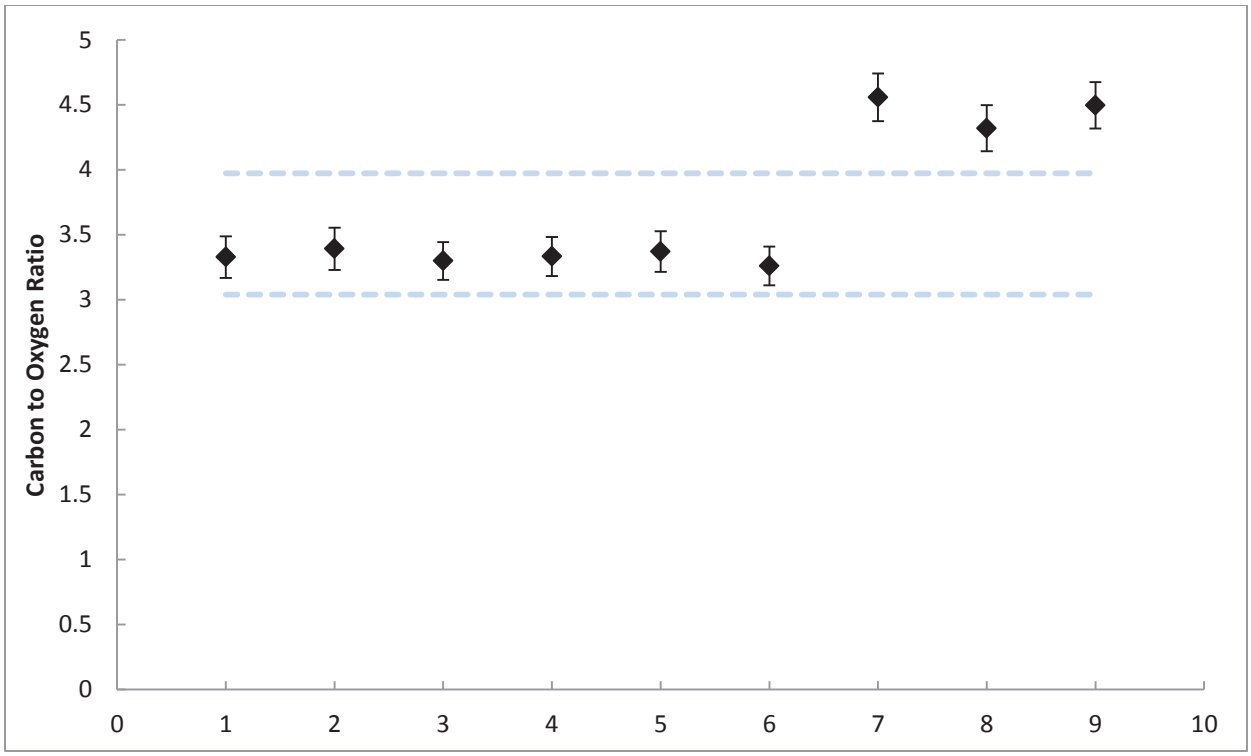

Figure 16. Carbon to oxygen ratio for simulated TNT.

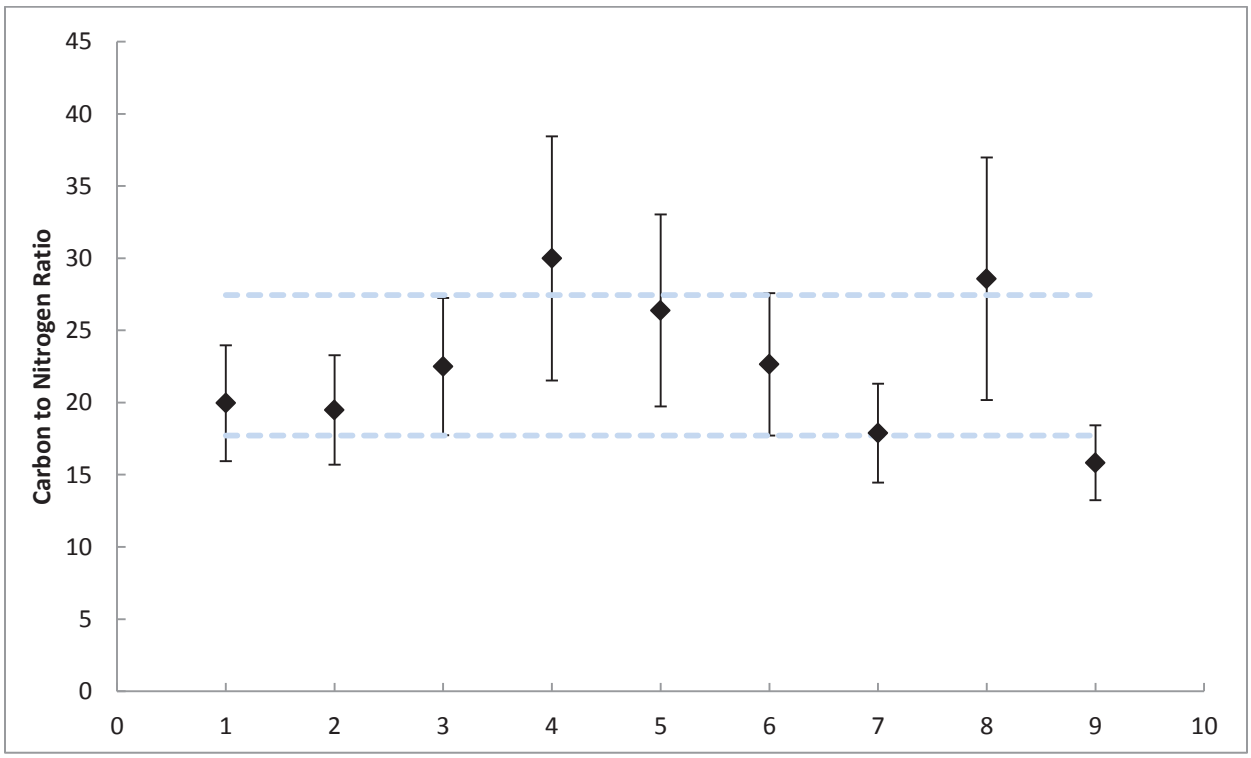

Figure 17. Carbon to nitrogen ratio for simulated TNT.

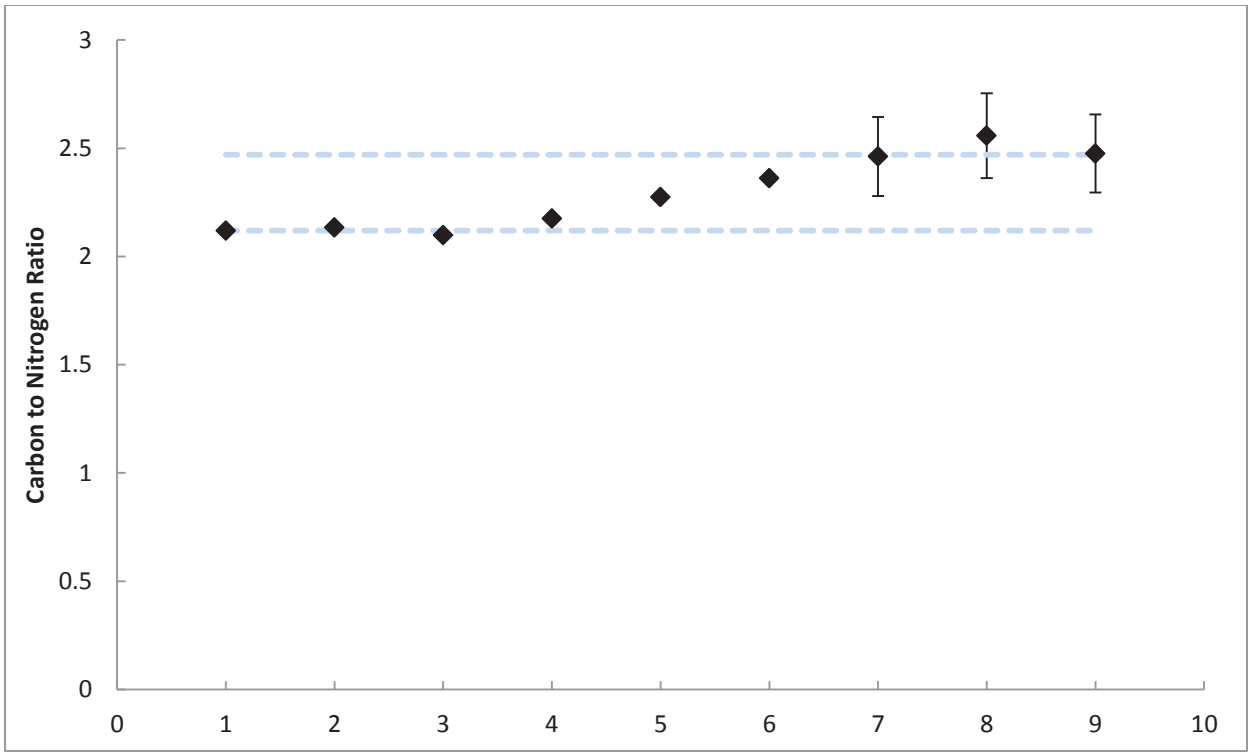

Figure 18. Carbon to nitrogen ratio for simulated TNT.

The previous figures are summarized in Table 2 below, which shows the average elemental ratios and their standard deviations for the three simulants. There are a few conclusions one can draw from the ratios and their standard deviations in Table 2. One of these is that those ratios that depend on the nitrogen inelastic scattering peak in general have a much higher variance than those that do not. This is to be expected given that the nitrogen peak area is dependent both on the area of the oxygen 6129 keV peak as well as the combination oxygen second-escape and nitrogen inelastic peak areas. A second observation is that the carbon to hydrogen ratio shows small variance in general. It would not be expected for this variance to remain small however if multiple geometries were used. Given that the carbon peak is generated through neutron inelastic scattering and the hydrogen peak through thermal neutron capture one would expect to see a significant dependence of this ratio on the geometry of the assessment.

Table 2 Elemental ratios for simulated explosives

	C/O	Stdev	C/N	Stdev	C/H	Stdev
HMX-sim	3.36	0.46	14.9	3.3	2.21	0.56
TATB-sim	5.57	0.47	10.7	3.3	1.87	0.19
TNT-sim	3.51	0.47	22.6	4.9	2.29	0.17

4. EFFECTS OF VARIANCE ON ASSESSMENT OF EXPLOSIVES

The important effect of variance in these measurements is of course how they affect the results of an assessment. If one were to assume that the only explosives identified were those three simulated explosives used in this study, one could then examine each individual measurement to determine how it would be assessed using a proposed algorithm. A simple example of such an algorithm would be to calculate the normal probability density function (pdf) for each measured ratio based on the average and standard deviations of the three different simulated explosives. One could then multiply these pdfs for each explosive type and determine which explosive type yields the highest net pdf. An example of this can be seen in Table 3 below, where the HMX C/N measurements are assessed based on the pdf for each elemental ratio when compared with each explosive type.

Table 3 Probability density functions for simulated HMX

Measurement #	C/N	TNT pdf	TATB pdf	HMX pdf
1	21.7 ± 11	0.0805	0.0004	0.0145
2	15.8 ± 6.4	0.031	0.0363	0.1154
3	13.4 ± 4.2	0.0136	0.087	0.1089
4	10.4 ± 2.4	0.0035	0.1204	0.0487
5	14.0 ± 4.2	0.0175	0.0727	0.1168
6	16.2 ± 5.8	0.0350	0.0296	0.1103
7	14.4 ± 4.3	0.0202	0.063	0.1195
8	11.2 ± 0.8	0.0054	0.1194	0.0663
9	16.5 ± 1.0	0.0373	0.0262	0.1068

As can be seen in Table 3, most of the measurements have the highest pdf when assessed against HMX. The only outlier is the first measurement where the TNT pdf is the highest. This is not surprising given that the uncertainty in this measurement's C/N ratio is approximately 50% of its value.

If we examine the explosives listed in Table 1, we can estimate the likelihood of the PINS system being able to distinguish each of them, based on calculations of their elemental ratios. Table 4 below shows the expected elemental ratios for these explosives based on MCNP calculations. If we assume that the C/O ratio for each explosive can have an expected variance of 15%, and the C/N ratio a 25% variance then we can make a better estimate of those explosives we can reasonably expect to distinguish from one another. From the table, we can see that we can reasonably expect to differentiate all the HMX and RDX-based explosives from the TATB-based explosives, i.e. PBX-9501 should be distinguishable from PBX-9502 given that their C/O and C/N ratios are different by approximately 40%. This is without making use of any other elements that might be present in the binders of these explosives, such as chlorine and fluorine in the Kel-F binding LX-17.

Table 4 Calculated Elemental ratios for explosives

Explosive	**C/O**	**C/N**	**C/H**
HMX	0.89	2.84	2.50
TNT	1.78	11.16	7.54
TATB	1.57	5.02	5.08
PBX-9501	0.95	3.19	2.48
PBX-9502	1.63	5.17	5.8
PBX-9404	0.92	3.06	2.78
LX-17	1.66	5.27	6.26
LX-10-0	0.97	3.10	2.65
Comp. B	1.24	4.87	4.11

5. CONCLUSIONS AND RECOMMENDATIONS

The variance of the elemental ratios used by the PINS system to identify explosives has been examined by analyzing a set of replicate measurements of both background and simulated explosives. The background measurements have been found to have relatively minor variations in peak areas, which should not add a large amount of variance in later measurements of elemental ratios.

The elemental ratios themselves have significant variance in the replicate measurements, on the order of 15% for the carbon-to-oxygen ratio and 25% for the carbon to nitrogen ratio. Even with these significant variances, a large number of explosives are readily distinguished from one another.

One area that would warrant future examination is the effect of varying the explosive mass and geometry on the elemental ratios. These variations were not examined in this work.

6. ACKNOWLEDGMENTS

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